



INVESTOR IN PEOPLE

**The Patent Office
Concept House
Cardiff Road
Newport
South Wales
NP10 8QQ**



I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

Signed

Andrew Gersey

Dated

14 March 2001

THIS PAGE BLANK (USPTO)

THE PATENT OFFICE

H

-8 OCT 1998

The Patent Office

080CT98 E395669-1 002877
F01/7700 0.00 - 9821856.3

Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road
Newport
Gwent NP9 1RH

1. Your reference CPR 50677/GB/P

2. Patent application number
(The Patent Office will fill in this part)

08 OCT 1998

9821856.3

3. Full name, address and postcode of the or of each applicant (underline all surnames)

IMPERIAL CHEMICAL INDUSTRIES PLC
IMPERIAL CHEMICAL HOUSE

MILLBANK

LONDON

SW1P 3JF

Patents ADP number (if you know it)

935003

If the applicant is a corporate body, give the country/state of its incorporation

UNITED KINGDOM

4. Title of the invention

BIPOLAR PLATES FOR FUEL CELLS

5. Name of your agent (if you have one)

Ieuan THOMAS

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

ICI GROUP INTELLECTUAL PROPERTY
INTELLECTUAL PROPERTY DEPARTMENT
PO BOX 11, THE HEATH
RUNCORN, CHESHIRE
WA7 4QE

Patents ADP number (if you know it)

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number
(if you know it)

Date of filing
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

YES

- a) any applicant named in part 3 is not an inventor, or
 - b) there is an inventor who is not named as an applicant, or
 - c) any named applicant is a corporate body.
- See note (d))

9. Enter the number of sheets for any of the following items you are filing with this form.

Do not count copies of the same document

Continuation sheets of this form

Description	7
Claim(s)	2 <i>80</i>
Abstract	0
Drawing(s)	1 <i>11</i>

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents (please specify)

11.

I/We request the grant of a patent on the basis of this application.
IMPERIAL CHEMICAL INDUSTRIES PLC

Signature *[Signature]*

Date

7 Oct 98

DULY AUTHORISED OFFICER

12. Name and daytime telephone number of person to contact in the United Kingdom

Kath Chester

01928 518093

Warning

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

Notes

- If you need help to fill in this form or you have any questions, please contact the Patent Office on 0645 500505.
- Write your answers in capital letters using black ink or you may type them.
- If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.
- If you have answered 'Yes' Patents Form 7/77 will need to be filed.
- Once you have filled in the form you must remember to sign and date it.
- For details of the fee and ways to pay please contact the Patent Office.

Bipolar Plates for Fuel Cells

The present invention relates to bipolar plates for fuel cells, to fuel cells comprising such plates and particularly to so-called proton-exchange membrane fuel cells.

5 A fuel cell is an electrochemical device in which electricity is produced without combustion of fossil fuel. In a fuel cell, a fuel, which is typically hydrogen, is oxidised at a fuel electrode (anode) and oxygen, typically from air, is reduced at a cathode, to produce an electric current and form by-product water. An electrolyte is required which is in contact with both electrodes and which may be alkaline or acidic, liquid or solid.

10 Heat and water are the only by-products of the electrochemical reaction in fuel cells. Accordingly, the use of such cells in power generation offers potential environmental benefits compared with power generation from combustion of fossil fuels or by nuclear activity.

In proton-exchange membrane fuel cells, hereinafter referred to for convenience 15 as "PEM" fuel cells, the electrolyte is a solid polymer membrane which allows transport of protons from the anode to the cathode and is typically based on perfluorosulphonic acid materials. The electrolyte must be maintained in a hydrated form during operation in order to prevent loss of ionic conduction through the electrolyte.

A PEM fuel cell typically comprises two electrodes, an anode and a cathode, 20 separated by a proton-exchange membrane electrolyte. At the anode, hydrogen fuel catalytically dissociates into free electrons and protons. The free electrons are conducted in the form of usable electric current through the external circuit with which the fuel cell is in electrical contact. The protons migrate through the membrane electrolyte to the cathode where they combine with oxygen from the air and electrons from the external 25 circuit to form water and generate heat. Individual fuel cells may be combined into assemblies to provide the amount of power required.

A PEM fuel cell assembly comprises a plurality of such individual cells. In a fuel cell assembly bipolar plates, also known as fluid flow field plates, play a significant role. The bipolar plate is fabricated with surface features, for example a series of 30 corrugations, which provide gas flow channels which ensure essentially even distribution of input gases over the electrode surfaces. The bipolar plate should have

high electrical conductivity as an ohmic loss in the plate will reduce the overall stack efficiency..

Bipolar plates constructed from metals, referred to therein as bipolar terminal grids, have been described by Douglas et al in US 3,134,696. Bipolar plates constructed from carbon/polymer composites, referred to therein as bipolar current
5 collectors-separators, have been described by Lawrence in US 4,214,969. Bipolar plates constructed from graphite, referred to therein as fluid flow field plates, have been described by Wilkinson et al in WO 95/16287.

We have now found that the electrical conductivity of bipolar plates can be
10 increased by coating them with a coating of an electrocatalytically-active material.

By "electrocatalytically-active material" we mean a material which where used as an electrode or coating therefor catalyses electrochemical reactions at high current densities at potentials close to the equilibrium potential as is more fully described by R Greef et al in "Instrumental Methods in Electrochemistry", Ellis Horwood, 1990 and by
15 D Pletcher et al in "Industrial Electrochemistry", Chapman and Hall, 1990.

According to the first aspect of the present invention there is provided a bipolar plate for fuel cells for (a) conducting current from the anode of one cell unit to the cathode of the adjacent cell unit and (b) distributing fluid characterised in that it comprises a substrate with a coating of an electrocatalytically-active material as
20 hereinbefore defined and has surface features, for example a series of corrugations, which provide gas flow channels which ensure essentially even distribution of input gases over the electrode surfaces.

According to a further aspect of the present invention there is provided a fuel cell assembly comprising

- 25 a) a plurality of cell units each of which contains a proton-exchange membrane separating the cell into anolyte and catholyte chambers and with an anode and a cathode on opposite sides thereof;
- b) a bipolar plate disposed between adjacent cell units;
- c) a current collecting terminal disposed at each end of the assembly ;
- 30 d) means to feed hydrogen fuel to the anolyte chambers of the cell; and

e) means to feed an oxygen-containing gas to the catholyte chambers of the cell characterised in that each bipolar plates comprises a bipolar plate according to the first aspect of the present invention.

5 The anode and cathode may be discrete components but are preferably provided as integral parts of a single unit as is more fully described in WO 95/16287.

In the fuel cell assembly according to the further aspect of the present invention the plurality of cell units are connected in series.

The fuel cell assembly according to the further aspect of the present invention is connected to an external circuit via the terminal current-collectors

10 Preferably the terminal current-collectors comprise bipolar plates according to the first aspect of the present invention.

Whereas the fuel cell assembly and bipolar plate according to the present invention are typically planar we do not exclude the possibility that they may be cylindrical or tubular.

15 We do not exclude the possibility that the fuel cell assembly and bipolar plate according to the present invention may be used in liquid electrolyte fuel cells such as phosphoric acid and direct methanol fuel cells.

The electrocatalytically-active coating of which the bipolar plate according to the present invention may be comprised is typically derived from a metal, metal oxide or
20 mixtures thereof from Group 8 of the Periodic Table of Elements, namely Fe, Co, Ni, Ru, Rh, Pd, Os, Ir and Pt.

Suitable electrocatalytically-active coatings comprising mixtures of platinum group metal and platinum group metal oxide are described in our EP 0,129,374.

25 Suitable electrocatalytically-active coatings comprising mixtures of ruthenium oxide, non-noble metal oxide and noble metal or oxide thereof are described in our EP 0,479,423.

Suitable electrocatalytically-active coatings comprising mixtures of cerium oxide and at least one non-noble Group 8 metal are described in our EP 0,546,714.

30 The electrocatalytically-active coating is preferably ruthenium oxide or mixtures of ruthenium oxide with at least one of PtO, Sb₂O₃, Ta₂O₅, PdO, CeO₂, Co₃O₄ or preferably mixture of RuO₂ with at least one of TiO₂, SnO₂, IrO₂.

Where the electrocatalytically-active coating comprises a mixture of ruthenium oxide and another oxide the content of the ruthenium oxide may be in the range 0 - 100 mol %, and typically 5 - 90 mol %.

5 The thickness of the electrocatalytically-active coating on the bipolar electrode may be in the range 0.5 - 100 gm^{-2} , and typically 1 - 90 gm^{-2} .

We do not exclude the possibility that the electrocatalytically-active coating may comprise an intermediate layer between the substrate and the outer layer. As examples of such intermediate layers may be mentioned *inter alia* the heat-treated niobium oxide layer and the tantalum layer described in EP 0,052,986 and EP 0,107,934 respectively.

10 Where the coating comprises ruthenium oxide it may comprise a plurality of different layers, for example a layer of $\text{RuO}_2/\text{TiO}_2$ and a layer of $\text{RuO}_2/\text{SnO}_2$.

The substrate of which the bipolar electrode is comprised is typically a metal chosen from Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zr, Nb, Ag, Pt, Ta, Pb, Al, or alloys thereof, preferably titanium or stainless steel. However, we do not exclude the possibility that the
15 substrate may be formed from a polymer-based material.

The electrocatalytically-active coatings may be applied to the substrate by, for example, painting of a solution of precursors followed by thermal decomposition, thermal spraying, screen-printing of metal alloy, Physical Vapour Deposition (PVD), CVD, electroplating, electroless plating or spray pyrolysis.

20 Application of a electrocatalytically-active coating comprising an outer layer of ruthenium oxide and non-noble metal oxide to a substrate by PVD is more fully described in our WO 95/05499.

Application of a electrocatalytically-active coating comprising an outer layer of ruthenium oxide and non-noble metal oxide to a substrate by thermal spraying is more
25 fully described in our WO 95/05498.

Application of a electrocatalytically-active coating comprising an outer layer of (a) cerium and/or cerium oxide and at least one non-noble Group 8 metal, or (b) platinum and/or platinum oxide and ruthenium and/or ruthenium oxide by PVD is more fully described in our WO 96/24705.

30 We do not exclude the possibility that electrocatalytic coating may comprise an intermediate layer between the substrate and outer layer. As examples of such

intermediate layers may be mentioned inter alia the heat-resistant niobium oxide layer and tantalum oxide layer described in our EP 0,052,986 and EP 0,107,934 respectively.

Hydrogen fuel for use in the fuel cell according to the present invention may be obtained from, for example, natural gas or methanol. Oxygen for use in the fuel cell according to the present invention may be obtained from air.

It will be appreciated that in the fuel cell assembly according to the further aspect of the present invention the components thereof may be provided with aligned ports, eg slots, to form a manifold to allow flow of fuel gas and oxidant gas from the means to feed such gases to the cell to the anodes and cathodes respectively.

We do not exclude the possibility that different coatings may be applied to different surfaces of the bipolar plate according to the present invention.

The present invention is illustrated by reference to the accompanying drawing which illustrates, by way of example only, a fuel cell assembly according to the present invention.

In the drawing, ion-permeable membranes (1) and (2) have cathode electrodes (3) and (4) respectively and anode electrodes, not shown, bonded to each of their major surfaces. A bipolar plate (5) according to the present invention, provided with surface features (6), is disposed between ion-permeable membranes (1) and (2) in contact with the electrode surfaces thereof. Terminal plates (7) and (8), provided with leads (9) and (10) for delivering electric current generated in the cell assembly to an external circuit, are disposed adjacent membranes (1) and (2) respectively.

In the assembly, membrane (1) is held firmly between terminal plate (7) and bipolar plate (5) so as to form an oxidant gas chamber (11) and a fuel gas chamber (12). In like manner, membrane (2) is held firmly between terminal plate (8) and bipolar plate (5) so as to form an oxidant gas chamber (13) and a fuel gas chamber (14).

Hydrogen fuel is supplied to the anodes in the fuel gas chambers (12) and (14) via fuel gas inlet conduit (15) and by-products removed via conduit (16).

Oxidant gas is supplied to cathodes (3) and (4) in the oxidant gas chambers (11) and (13) via oxidant gas inlet conduit (17) and by-products removed via conduit (18).

Openings (19) and (20) located in opposite corners of membranes (1) and (2) are aligned with hydrogen gas inlet and outlet conduits (15) and (16) and with openings (21)

and (22) in bipolar plate (5) to facilitate passage of hydrogen fuel gas into the fuel chambers (12) and (14) and to remove by-products therefrom.

Openings, not shown, and openings (23) located in opposite corners of membranes (1) and (2) are aligned with oxidant inlet and outlet conduits (17) and (18) and with opening (24) and another not shown in bipolar plate (5) to facilitate passage of oxidant gas into the oxidant chambers (11) and (13) and to remove by-products therefrom.

End plates (7) and (8), membranes (1) and (2) and bipolar plate (5) are each provided with a plurality of openings (25) through which assembly tie-rods or bolts (26) pass.

In a further embodiment of the present invention, a layer of diffusion material which is electrically conducting and porous, for example a carbon-coated paper or a graphite-impregnated polymer film, is disposed in the oxidant gas chambers (11) and (13) and/or in the fuel gas chambers (12) and (14). For example, the layer of diffusion material may be disposed between polar plate (5) and the adjacent electrode surfaces of membranes (1) and (2) and/or between the terminal plates (7) and (8) and the adjacent electrode surfaces of membranes (1) and (2).

The present invention is further illustrated by reference to the following Examples.

Examples 1 and 2

A coating of composition 47.5 mole % ruthenium and 52.4 mole % titanium was prepared by adding tetra-n-butyl titanate (7.47g) to a solution of ruthenium (2g), as ruthenium trichloride, in pentan-1-ol (31g).

In Example 1, a portion of this solution was applied by brush to a titanium substrate which had been etched in 10% oxalic acid solution at 85°C for 8 hours. The coated substrate was dried at 180°C and then baked at 450°C; 12 coats were applied in this manner. Three of the coated plates were operated as bipolar plates in a PEM fuel cell and the cell voltage output thereof was determined at 1 A cm²/V and the per-centage volume efficiency calculated therefrom.

In Example 2 the procedure of Example 1 was repeated except that the substrate was a 316L stainless steel plate which had been grit blasted.

In a Comparative Test, stainless steel plates were operated as bipolar plates in a PEM fuel cell.

The results are shown in the Table from which it can be seen that PEM fuel cells according to the present invention have a voltage efficiency at least 13% better than a
5 PEM fuel cell comprising a known plate.

TABLE

Bipolar plate material	Voltage efficiency %
316L SS	100
Example 1	113
Example 2	117

Claims

1. A bipolar plate for fuel cells for (a) conducting current from the anode of one cell unit to the cathode of the adjacent cell unit and (b) distributing fluid characterised in that it comprises a substrate with a coating of an electrocatalytically-active material as
5 hereinbefore defined and has surface features, for example a series of corrugations, which provide gas flow channels which ensure essentially even distribution of input gases over the electrode surfaces.
2. A bipolar plate for fuel cells as claimed in Claim 1 further characterised in that the substrate is metallic.
- 10 3. A bipolar plate for fuel cells as claimed in Claim 1 further characterised in that the electrocatalytically-active material is derived from one or more platinum group metals or oxides thereof, ie Pt, Rh, Ir, Ru, Os or Pd, or cerium.
4. A bipolar plate for fuel cells as claimed in Claim 3 further characterised in that the electrocatalytically-active material is derived from ruthenium or an oxide thereof.
- 15 5. A bipolar plate for fuel cells as claimed in Claim 4 further characterised in that the electrocatalytically-active material comprises ruthenium oxide and a non-noble metal oxide.
6. A bipolar plate for fuel cells as claimed in Claim 5 further characterised in that the electrocatalytically-active material comprises a noble metal or oxide thereof.
- 20 7. A bipolar plate for fuel cells as claimed in Claim 4 further characterised in that the electrocatalytically-active material comprises mixtures of RuO_2 with at least one of TiO_2 , SnO_2 , IrO_2 , PtO , Sb_2O_3 , Ta_2O_5 , PdO , CeO_2 , Co_3O_4 .
8. A bipolar plate for fuel cells as claimed in Claim 7 wherein the electrocatalytically-active material comprises $\text{RuO}_2/\text{TiO}_2$.
- 25 9. A fuel cell comprising
 - a) at least two bipolar plates ;
 - b) a membrane assembly disposed between the plates, which membrane assembly comprises a pair of opposed electrodes with a hydraulically impermeable proton-exchange membrane disposed therebetween with the proviso that where the fuel
- 30 cell comprises more than two bipolar plates a membrane assembly and a bipolar plate

alternate throughout the cell and the membrane assemblies are disposed in the fuel cell such that an anode and a cathode alternate throughout the cell;

c) means to feed gaseous hydrogen fuel to the anodes; and

d) means to feed an oxygen-containing gas to the cathode,

5 characterised in that each bipolar plates comprises a bipolar plate as claimed in Claim 1.

10. A fuel cell assembly comprising

a) a plurality of cell units ; and

b) at least one bipolar plate as claimed in Claim 1 disposed between adjacent cell units.

11. A fuel cell assembly comprising

10 a) a plurality of cell units wherein each unit contains a gas impermeable proton-exchange membrane separating the cell into anolyte and catholyte chambers and with an anode and a cathode on opposite sides thereof;

b) a bipolar plate disposed between adjacent cell units;

c) a current collecting terminal disposed at each end of the assembly ;

15 d) means to feed hydrogen fuel to the anodes; and

e) means to feed an oxygen-containing gas to the cathode,

characterised in that each bipolar plates comprises a bipolar plate as claimed in Claim 1.

20

25

30

THIS PAGE BLANK (USPTO)

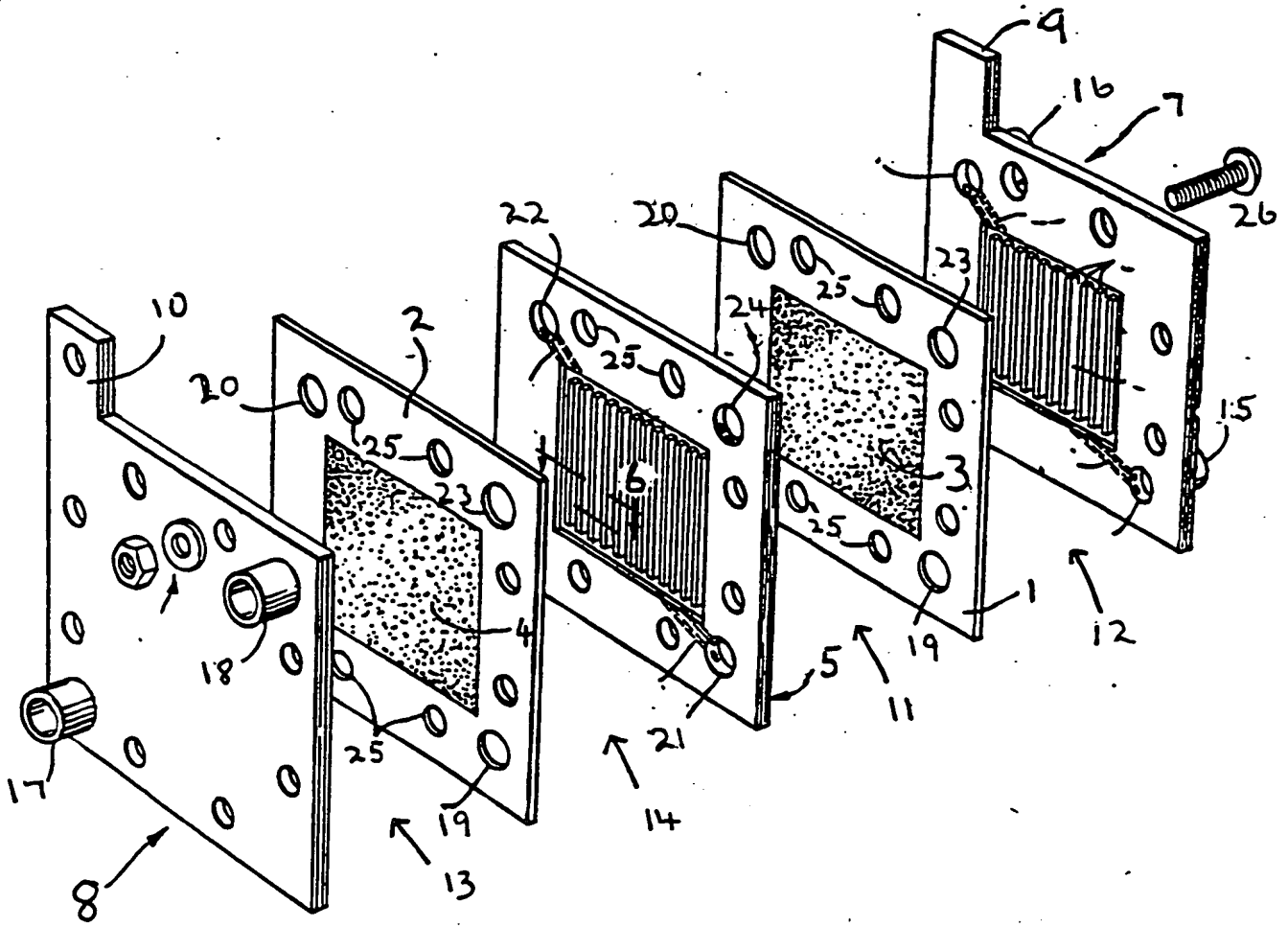


FIG. 1

MAY ET AL

Serial No. 09/805,145

Filed: March 14, 2001

FUEL CELLS AND FUEL CELL PLATES

THIS PAGE BLANK (USPTO)